

Domain excitations in spin-Peierls systems.

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We study a model of a Spin-Peierls material consisting of a set of antiferromagnetic Heisenberg chains coupled with phonons and interacting among them via an inter-chain elastic coupling. The excitation spectrum is analyzed by bosonization techniques and the self-harmonic approximation. The elementary excitation is the creation of a localized domain structure where the dimerized order is the opposite to the one of the surroundings. It is a triplet excitation whose formation energy is smaller than the magnon gap. Magnetic internal excitations of the domain are possible and give the further excitations of the system. We discuss these results in the context of recent experimental measurements on the inorganic Spin-Peierls compound CuGeO_3 .

The recent discovery of the first inorganic Spin-Peierls (SP) compound CuGeO_3 has renewed the interest in the subject of quasi-one-dimensional spin-phonon coupled systems¹. In spite of the intense experimental and theoretical activity devoted to the study of this system neither the excitations in the low temperature phase nor the mechanism of the Spin-Peierls transition itself are well understood yet. Most studies have used as a model Hamiltonian a Heisenberg antiferromagnetic chain with alternating coupling and next nearest neighbors (nnn) interaction^{2,3}. The Spin-Peierls transition is supposed to arise from a competition between the elastic energy cost necessary to dimerize the chain and the magnetic energy gain in this process. The dynamics of the phonons are then supposed to be independent of the magnetic subsystem and an extreme adiabatic approximation is assumed.

In spite of the success to reproduce both thermodynamical as well as dynamical properties of Spin-Peierls systems by this approach, there are now several indications that the Spin-lattice interaction in CuGeO_3 is strong enough to make insufficient this approximation. No soft phononic mode related to dimerization has been found⁴. This fact points toward an order-disorder type transition where nonlinear excitations (domain wall) different from phonons drive the structural transition. Otherwise, the phase diagram of the system in presence of a magnetic field is now well established. Above a critical value of the magnetic field the system undergoes a transition from

the uniform dimerized into an incommensurate phase. The incommensurate lattice pattern has been measured by X-ray experiments and interpreted as soliton lattice structure⁵. The role of solitons in CuGeO_3 was indeed previously emphasized⁶. They could give a unified picture of the above features. They could also be relevant to explain the rapid reduction of the SP temperature by doping and the apparition of an antiferromagnetic phase^{6,7}.

It seems then natural to build the excitation spectrum of SP systems on the basis of some kind of solitonic excitation (in this work we use the term soliton in the general sense of a finite energy localized configuration which is simultaneously magnetic and structural excitations, we call kinks the 1D solitons). The present work gives some insight in this direction.

Moreover, recent neutron scattering studies have given a detailed information about magnetic excitations in CuGeO_3 ⁸. They show a 'double gap' structure, in addition to a dispersive triplet excitation there is another gap which separates the first peaks from the band edge of a continuum. This continuum has been interpreted as due to kink excitations⁸. It was stated that no free kinks could exist due to the inter-chain coupling⁹. The inter-chain coupling were taken into account in previous works as providing a linear confined potential between the kinks⁶. This potential arises from a mean field approximation to the inter-chain coupling. Particularly in a recent work a ladder of bound kink-antikink states was shown for this model¹⁰.

In this paper we study the formation of solitonic structures in SP system including the three dimensional character of the phonon field. We use bosonization techniques to account for the low energy magnetic excitations. We show that the excitation gap of the system corresponds to create a domain-like localized structure in which the dimerization is in antiphase to the one in the bulk material. No independent kinks are possible. Instead of that the walls of the domain always find an equilibrium situation depending on the value of the inter-chain interaction. Internal excited states of the domain are the higher excited levels of the system. They could be connected with the continuum seen in CuGeO_3 .

Let us consider a system of Heisenberg antiferromagnetic chains immersed in the phonon field of the mate-

rials. The spin-phonon coupling arises from the modulation of the magnetic exchange by the lattice motion. Therefore we will focus on the following spin-phonon Hamiltonian:

$$H = H_{ph} + H_{mg} \quad (1)$$

$$\frac{H_{ph}}{J} = \sum_{i,j} \frac{P_i^j{}^2}{2M} + \frac{K_{\parallel}}{2}(u_{i+1}^j - u_i^j)^2 + \frac{K_{\perp}}{2}(u_{i+1}^{j+1} - u_i^j)^2 \quad (2)$$

$$\frac{H_{mg}}{J} = \sum_{i,j} (1 + (u_{i+1}^j - u_i^j)) \mathbf{S}_i^j \cdot \mathbf{S}_{i+1}^j \quad (3)$$

we denote by i the site in a j -th chain. \mathbf{S}_i^j are spin-1/2 operators of the i, j ion. H_{ph} represents our simplified model for the phonons. It contains a scalar coordinate u_i^j and its conjugate momentum P_i^j which are supposed to be the relevant ion coordinates for the dimerization process. Notice that, in CuGeO_3 , these are mainly related with the displacement of the O ions. K_{\parallel} and K_{\perp} are the longitudinal and transversal elastic couplings. We have included only the coupling in one direction, generalization to the other direction is straightforward and produces only a redefinition of the quantity $\mathbf{B}(j)$ of Eq. (12). This transversal coupling gives the three dimensional character to the phonons field which has been neglected in previous studies of solitons in SP system¹¹. It is essential for our considerations. The model Hamiltonian given by (1) does not include magnetic interaction between the chains. We will discuss latter on the possible effect of this interaction on the excitation spectrum. Moreover we have not introduced a nnn interaction between the spins of a chain. Even though, it play an important role in reproducing several experimental properties in CuGeO_3 we do not expect any qualitative change of the results presented below which are based on the solitonic excitations and not strongly affected by this frustrated interaction^{13,14}.

A similar model of the one given by Eq. (1) were recently studied by Essler, Tsvelik and Delfino¹⁵ using rigorous field theoretical results. However, as the authors of ref. 15 have integrated out the lattice coordinates and neglected the dynamic of the phonons they did not take into account the mixed excitation we will discuss in the present paper. We remark that in CuGeO_3 the scale of magnetic and phononic excitation are of the same order of magnitude suggesting that the elimination of the phonons degree of freedom is not suitable.

Owing to the one-dimensional character for the magnetic interaction in (1) we can resort to the bosonization techniques to take into account the low energy excitations of the systems¹⁶. The spin variables are approximately represented by the bosonic fields $\phi^j(x)$ and its conjugated momentum $\Pi^j(x)$. In addition we retain only the phonon

modes producing a smooth deviation of the dimerized pattern. So, we make the replacement $(-1)^i u_i^j \rightarrow u^j(x)$. The effective Hamiltonian ($H_{eff} = H_{ph} + H_{mg}$) is given by:

$$H_{ph} = \sum_j \int dx \left\{ \frac{P^j(x)^2}{2M} + 2K_{\parallel} u^j(x)^2 + \frac{K_{\perp}}{2} (u^{j+1}(x) - u^j(x))^2 \right\} \quad (4)$$

$$H_{bos} = \sum_j \int dx \left\{ \frac{1}{2\pi} \left[\frac{v_s}{\eta} (\partial_x \phi^j(x))^2 + v_s \eta (\pi \Pi^j(x))^2 \right] + u^j(x) \sin \phi^j(x) \right\} \quad (5)$$

v_s is the spin wave velocity and η the exponent of the correlation functions^{16,11}. For the isotropic Heisenberg model with nn interaction we have $v_s = \pi/2$ and $\eta = 2$.

We analyze the excitations of H_{eff} by using a self-consistent harmonic approximation (SHA) which has been shown to be reliable for Spin-Peierls systems¹¹. It is nearly connected to the semi-classical methods widely used in the study of nonlinear field theory¹². In the SHA the fields ϕ^j are split into a classical variable ϕ_0^j and its quantum fluctuation $\hat{\phi}^j$. Terms up to quadratic order in $\hat{\phi}^j$ are retained and the annulment of the first order term is required. The resulting equations are:

$$-\frac{2v_s}{\eta} (\partial_x^2 \phi_0^j) + u^j e^{-\langle \hat{\phi}^j \rangle / 2} \cos \phi_0^j = 0 \quad (6)$$

$$\sin \phi_0^j e^{-\langle \hat{\phi}^j \rangle / 2} + (2K_{\perp} + 4K_{\parallel}) u^j + K_{\perp} (u^{j+1} + u^j) = 0 \quad (7)$$

$\langle \hat{\phi}^j \rangle$ is the ground state expectation value and the last equation is the classical equation for the lattice coordinates u^j . Uniform solutions of (6) and (7) correspond to the homogeneous dimerized configuration of the chains. They are given by:

$$\phi_0^j = \frac{\pi}{2} (\text{mod}. 2\pi) \quad , \quad u^j(x) = u_0 = \frac{1}{\pi(4K_{\parallel})^{3/2}} \quad (8)$$

the transverse coupling is not effective in this case and the state is the same as the one of a single chain problem. Excitations over this state are given by:

$$\omega(k) = v_s (k^2 + k_0^2)^{1/2} \quad (9)$$

they represent a band of triplet excitations separated from the ground state by the gap $\Delta = v_s k_0 = (\pi u_0)^{2/3}$. These magnon-like excitations are usually characterized as the elementary excitations of a Spin-Peierls system.

However, there is a lower excitation energy of the system which arises when inhomogeneous solutions of (6) and (7) are allowed. To take it into account we invert

eq. (7) to obtain $u^j(x)$ and then replace it in eq (6). The result is:

$$u^j(x) = u_0 \sum_{j'} \mathbf{B}(j' - j) \sin \phi_0^{j'}(x) \quad (10)$$

$$\partial_x^2 \phi_0^j + \frac{1}{\xi^2} \sum_{j'} \mathbf{B}(j' - j) \cos \phi_0^j \sin \phi_0^{j'} = 0 \quad (11)$$

with:

$$\mathbf{B}(j' - j) = \int_{-\pi}^{\pi} \frac{dk}{(2\pi)} \frac{\cos(k(j' - j))}{1 + \epsilon \sin^2 \frac{k}{2}} \quad (12)$$

$\epsilon = \frac{K_{\perp}}{K_{\parallel}}$ is the relative inter-chain elastic couplings and $\xi = v_s/\Delta$ will become the characteristic width of the domain wall (see below).

Eq. (10) and (11) are our basic system of differential equations to search solitonic excitations of the system (each equation is labeled by the index j of the chain). They should be solutions of the system under the requirement of total finite energy respect to the uniform configuration. The inter-chain elastic coupling strongly limits the form of these solutions. For example, free kinks do not fulfill this condition because they create an infinite zone where the transverse couplings are active. The fields $u^j(x)$ and $\phi^j(x)$ should go as $x \rightarrow \pm\infty$ to the values they have in the uniform configuration (8). The simplest solution satisfying these conditions can be constructed as follows. We freeze the value of ϕ_0^j in all the chains but the 0-th to its value in the homogeneous SP state, i.e. $\phi_0^j = \pi/2$ for $j \neq 0$. The system (11) then reduce to a simple equation over the 0-chain. It corresponds to the classical equation for an effective one-chain problem with renormalized parameters and an additional term favoring the dimerization phase of the neighbors chains. We solve the equation by direct integration imposing the previously discussed boundary conditions. The result is:

$$t(x) \equiv \sin(\phi_0^0) = 1 - 2 \frac{\cosh^2[x_0/\xi] \operatorname{sech}[(x - x_0)/\xi]}{\operatorname{sech}[(x + x_0)/\xi]} \quad (13)$$

with

$$x_0 = \frac{1}{2} \log \left[\frac{1 + \mathbf{B}(0) + 2\sqrt{\mathbf{B}(0)}}{1 - \mathbf{B}(0)} \right] \quad \mathbf{B}(0) = 1/\sqrt{1 + \epsilon} \quad (14)$$

This solution is shown in fig. (1). We call it a domain. $2x_0$ measures the size of the domain configuration (see the insertion of fig. (1)). It goes to infinity as the chain get decoupled ($\epsilon \rightarrow 0$). In this limit, expression (13) becomes a product of two tanh-kink form. A small ϵ produces a rapid accommodation of the walls one near the other in order to reduce the inter-chain energy. For a greater ϵ , $2x_0$ departs from the distance between the zeros of $t(x)$ called δ in the inserted figure. As the walls

could not collapse this value never reaches the double of the width of the wall (2ξ). Note that for each inter-chain coupling we have an equilibrium distance. We do not find a linear confinement potential between the kinks as it is assumed in other analysis^{6,10} of this problem. The domains turn out to be the elementary excitations of the system. They will move as a whole when the translational invariance be restore. This is a triplet excitation because its total S_z could be 0 or ± 1 . Note that in the bosonic representation the total magnetization is given by $S_z^{tot} = \frac{1}{2\pi} \sum_j (\phi^j(\infty) - \phi^j(-\infty))$.

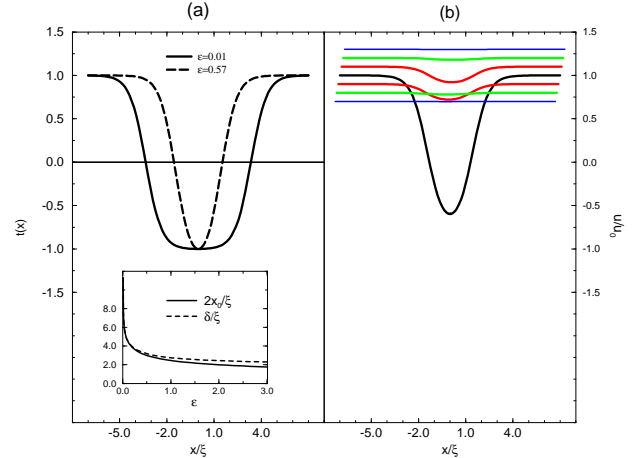


FIG. 1. (a) function $t(x)$ defined in (13). It gives the magnetic profile of the domain excitation. The insertion shows the ϵ -dependence of the parameter $2x_0$ (related to the domain's size) and δ named the distance between the zeros of $t(x)$. b) Lattice pattern corresponds to (15) for the $\epsilon = 0.57$. We show the deformation of the 0-chain and its neighbours chains.

The lattice deformation pattern is obtained from (10):

$$u^j(x) = u_0 \mathbf{B}(j) \sin \phi_0^0 + (1 - \mathbf{B}(j)) u_0 \quad (15)$$

We show this configuration in fig. (1b). The displacement amplitude in the intermediate zone of the 0-th chain is smaller than u_0 . The distortion is not restricted to the 0-chain as for the bosonic field ϕ_0 . The neighbors chains are slightly distorted respect to the uniform dimerized pattern. This distortion is indeed very weak as the result of the rapid fall of the value of $\mathbf{B}(j)$ with j ¹³. This behavior together with the asymmetry seen between the magnetic and lattice pattern is possible as a consequence of neglecting the inter-chain magnetic interaction. We should expect that inclusion of this interaction will smear out the deformation producing a kind of two (or three) dimensional domain with dimerization pattern opposite to the one of the surrounding material. Note that in our model spin-one excitations live in a chain but on turning off the transversal magnetic exchange a triplet localized state could appear exciting simultaneously several chains. Summarizing, a general solution of (11) in presence of a

magnetic interaction will be an enlarged domain in the direction of the chains.

We now turn to the study of the quantum states of the theory in terms of our classical solution. The first contribution comes from the creation energy of the domain. It is the difference of the values of the classical energy plus the zero point energy of the fields obtained with and without the domain. The classical energy is evaluated by replacing (13) and (15) in (4) and (5) and subtracting the energy of the uniform configuration. After a long but straightforward calculation we obtain:

$$E_{cl} = -\frac{1}{8\pi K_{\parallel}} [\mathbf{B}(0)I_2 + 2I_1] \quad (16)$$

with:

$$I_1 \equiv \frac{4}{\sqrt{\mathbf{B}(0)}} x_0$$

$$I_2 \equiv -\frac{2}{\sqrt{\mathbf{B}(0)}} \left[\frac{2}{\sqrt{\mathbf{B}(0)}} - 2\left(1 + \frac{1}{\mathbf{B}(0)}\right)x_0 \right]$$

The ϵ -dependence of this classical energy is shown in fig. (2a). For $\epsilon \rightarrow 0$ we have $E_{cl} = \frac{1}{2\pi K_{\parallel}}$ which is twice the classical creation energy of a kink in the single chain problem (expressions (4.10) and (4.11) of ref. 11 with the appropriate redefinitions of the constants). When the inter-chain coupling is switch on, the walls get closer and its total energy slightly increases. Note that in fig.(2a) we are showing the energies in units of the magnon gap Δ , therefore for any reasonable choice of ϵ it will be more favorable to create a domain rather than to excite a magnon. This result is at the hearth of our claim that the domains are the lower energy excitations of SP systems and will survive when quantum fluctuation are included.

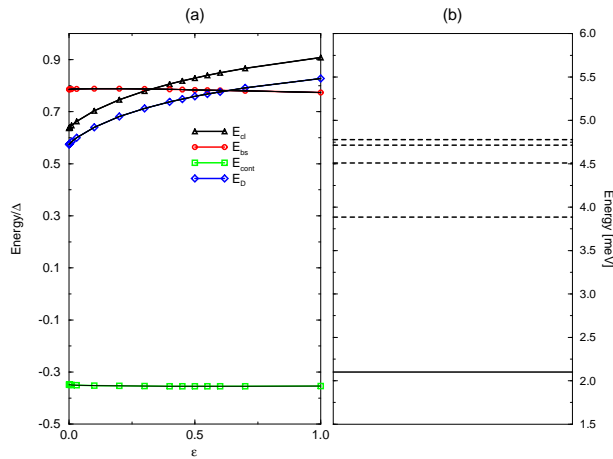


FIG. 2. (a) Differents contribution to the domain energy (E_d). E_{cl} classical energy, E_{bs} contribution of the bound state of the potential (17), E_{cont} contribution of the continuu(see text). (b) Energy levels diagram for the parameter fitted for CuGeO₃. Dashed lines denote the excited levels of the domain

We now go to the inclusion of these quantum fluctuations. The fluctuation operator is the quadratic form in the fields $\hat{\phi}^j$ of the SHA Hamiltonian. Its eigenvalue problem corresponds to a set of Schroedinger-like equations for the following potentials:

$$V^0(x) = (\mathbf{B}(0)(t(x) - 1) + 1)t(x) \quad (17)$$

$$V^j(x) = \mathbf{B}(j)(t(x) - 1) + 1 \quad (18)$$

Once the eigenvalues (E_λ) are known the frequency oscillations in presence of the domain could be computed as $\omega_d(\lambda) = \Delta\sqrt{E_\lambda}$. The quantum correction to the classical energy (16) is the difference between the sum of the zero point energies of these oscillators and the ones given by (9) i.e. in absence of the domain (the cutoff dependence is eliminated when the term containing $\langle \hat{\phi}^j{}^2 \rangle$ in the SHA Hamiltonian is included¹¹).

Equation (17) defines a quantum mechanics problem of a particle in a double well potential, whereas, Eq. (18) the one of weak wells. We have solved numerically the eigenvalues problem. The bound states has been obtained by means of a nodes counting algorithm. Numerical details of the method will be given elsewhere. For the double well potential (17), we have found two bound states (called E_1 and E_2). For $\epsilon \rightarrow 0$, the wells get far apart and the two levels collapse in a degenerate bound state with the same energy as the bound state of the single kink problem¹¹. The larger ϵ , the bigger is the splitting due to the tunneling between the wells.

Potentials (18) have only one bound state near the border of the continuum due to the weakness of the potential. We neglect its contribution to the total creation energy as well as the slightly distortion of the continuum. We will discuss later on the relevance of these bound states as excited levels of the domain.

The contribution of the continuum levels of (17) could be computed once the phase shift $\delta(k)$ of these states is known. We calculate its by numerical integration of the equation starting from the origin and matching its behavior to its asymptotic behavior at $x \rightarrow \infty$. The final expression for the total contribution of these states is:

$$E_{cont} = \frac{\Delta}{2} \left\{ \frac{1}{\pi} \int_0^\infty \frac{2 - k\delta(k)}{\sqrt{k^2 + 1}} dk - 1 \right\} \quad (19)$$

The last integral has been numerically evaluated. In fig. (2a) we show the ϵ -evolution of the different pieces contributing to the total creation energy (E_d). The main dependence comes from the classical energy. We find, again, that in the limit of decoupled chains our excitation energy corresponds to the total energy of two independent kinks¹¹.

What about CuGeO₃? As we have stated in the introduction, neutron scattering experiments show a clear dispersive excitation at 2.1 meV. This excitation was previously analyzed on the basis of a dimerized and frustrated

Heisenberg chain^{9,17,18}. Taking into account the previous results we associate the energy gap with the energy formation of a domain. To fix parameters we use the experimentally fitted values $J = 120\text{K}$ and $u_0 = 0.042$. With these values we have $\Delta = 2.68\text{ meV}$ larger than the gap. We can therefore choose $\epsilon = 0.57$ giving E_d equal to the measure gap. For this ϵ the walls are near and the domain seems as a local depression of the SP order (see fig.1). Inclusion of nnn interaction will change the parameters. The next energy levels of the system are internal excitations of the domain. They correspond to the bound states previously discussed. Fig. (2b) shows a level diagram for the low energies excitation of the system. It is natural to associate the transitions to the excited levels with the continuum seen in CuGeO_3 . We recall that switching on the inter-chain magnetic coupling will smoother of the domain deformation in the direction perpendicular to the chains. Therefore we expect that the internal excitations will form a continuous band. Note also that a two domain continuum is possible starting near $2E_d$. To give precise predictions of the effect of the domain formation on the neutron scattering spectra the detailed information about the dynamics correlation functions will be needed. Note in addition that as our domain is referred to a given state of dimerization degeneration between the states at $q = 0$ and $q = \pi$ is expected.

Let us assume a level excitations diagram for CuGeO_3 as the one of fig. (2b). As a finite density of domain will be thermally created our model predict transitions corresponding to rise the domain to its excited levels. The first one should appear as a low energy peak at about $\omega_d(1) = 1.78\text{ meV}$ in the optical response of the system and to increase its intensity with the temperature up to T_{SP} . Recent inelastic light scattering experiences¹⁹ have identify a new resonance in agreement with this prediction. However, note that this transition were assigned in Ref 19 to a three-magnon process on the externally dimerized chain. More experimental and theoretical work will be need to decide between these interpretations.

Finally, we have discussed in this paper only the magnetic excitations of the system. However the domain is a mixed state between the spins and phonons. So we expect that the phonon response will be sensitive to the domain formations. The apparition of shoulder or satellite peaks related to the frequency oscillations of the ions around the domain should be expected²⁰.

In summary, we have identified a new excitation mode of Spin-Peierls systems. It gives a natural extension of the usual singlet-triplet excitation of the statically dimerized chain when the lattice relaxation is allowed. As the excited spins are not confined to a dimer, internal excitation are possible. We claim that these excitations could account for some of the features seen in recent experiments.

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